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Charged and Neutral Binary Nucleation of Sulfuric Acid in Free Troposphere Conditions

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Abstract. We present a data set of binary nucleation of sulfuric acid and water, measured in the CLOUD chamber at CERN during the CLOUD3 and CLOUD5 campaigns. Four parameters have been varied to cover neutral and ion-induced binary nucleation processes: Sulfuric acid concentration (1e5 to 1e8 molecules per cm^{-3}), relative humidity (10% to 80%) , temperature (208-293K) and ion concentration (0-4000 ions per cm^{-3}). In addition, classical nucleation theory implemented with hydrates and ion induced nucleation is compared with the data set. Our model and data are also compared with nucleation rates measured at Puy de Dome in the tropopause.

Keywords: binary nucleation, ion-induced nucleation, CLOUD, Free Troposphere.
PACS: 92.60.Mt, 82.60.Nh .

INTRODUCTION

Up to half of the aerosol acting as cloud condensation nuclei originates from aerosol nucleation[1]. The possible influence of cosmic rays on aerosol nucleation is of considerable interest[2].The CLOUD experiment at CERN aims to study, under controlled conditions, nucleation processes as well as the effects of ions on nucleation. CLOUD was installed at CERN in 2009 and seven successful campaigns were carried out to study ion-induced and neutral binary nucleation of H_2SO_4 and H_2O, for tropospheric and stratospheric conditions. In this presentation, we will present the nucleation data obtained in the CLOUD chamber together with the new model which includes an ion-induced nucleation mechanism. To complete the picture, first molecular observation of nucleation in the tropopause will be shown and compared with the CLOUD data and the newly improved classical nucleation theory.

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LABORATORY MEASUREMENT

Technical input for the CLOUD design was obtained in a pilot experiment in 2006 [3].The chamber is a 3m-diameter electro-polished stainless-steel cylinder (26.1 m^3). A field cage is installed inside the chamber to allow the removal of ions, when required. The contents of the chamber are irradiated by UV light in the range 250-400 nm. Experimental runs can be performed at stable temperatures between 40°C and -65°C.

The chamber is exposed to a 3.5 GeV/c secondary pion beam from the CERN PS, corresponding to the characteristic energies and ionization densities of cosmic ray muons in the lower troposphere. The beam intensity can be adjusted to cover the natural range from ground level to the stratosphere. Ultra-pure air is obtained from the evaporation of cryogenic liquid N_2 and liquid O_2. The air is humidified with a Nafion humidifier. Ozone is added to the air by UV irradiation. Trace gases such as SO_2 are added from gas cylinders containing pressurised N_2 as the carrier.

The chamber instrumentation includes PTRMS, CIMS, Nano-SMPS, CPC battery, PSM, API-ToF, NAIS, Gerdien, LOPAP, dew point sensor, SO_2 and O_3 analyser, as well as T, P and UV sensors. Sulphuric acid was generated by means of UV light, SO2 and water.

From the API-ToF data, pure binary nucleation of sulphuric acid and water can be clearly identified[4]. Nucleation processes for species other than water and sulphuric acid have been excluded from this study.

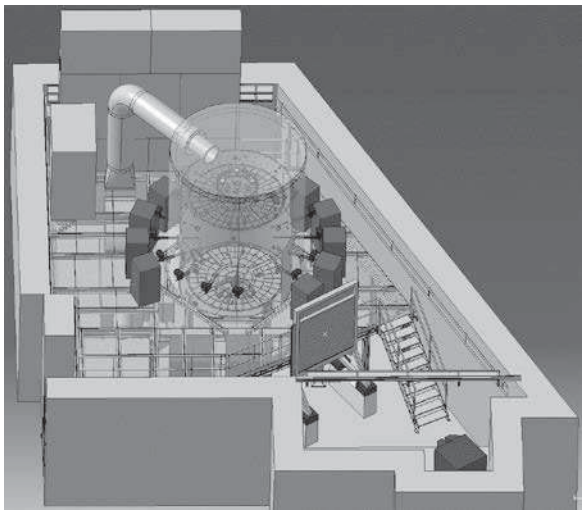

FIGURE 1. An illustration of CLOUD in the T11 experimental zone at the CERN PS. The de-focused particle beam exits a dipole magnet (bottom right), crosses the aluminium hodoscope counter (middle) and then traverses the 3m-diameter CLOUD chamber, before being stopped by the concrete wall (top left). The instruments (blue boxes) analyse the contents of the chamber via sampling probes. The temperature of the air is precisely controlled and circulates between the chamber and the thermal housing. The clearing field for the removal of ions is shown by transparency (bottom and upper part of the chamber). A technical description can be found in the supplementary of Kirkby et al. [4].

MODEL

The classical nucleation theory including the hydrate interaction model and the ion-induced nucleation has been merged into one model. From this improved model an extension of the parameterisation from Vehkamäki et al. [5] has been developed, and compared with the CLOUD and Puy de Dome data.

FIELD MEASUREMENT

To complete this study, a field measurement at the high altitude research station, Puy de Dome has been performed during winter 2012 (Figure 2). The puy de Dôme research station is located at 1 465m above sea level in central France (45:460 N, 2:57 0E). A detailed description of the site is provided by Venzac et al. [6]. The Puy de Dôme research site contains a number of different instruments capable of resolving the physical, chemical, optical, and hygroscopic properties of aerosol particles. During this campaign nucleation processes have been measured with an API-ToF, NAIS and PSM. Comparison of these nucleation events with model and cloud data will be presented.

FIGURE 2. Unfreezing the inlets at the Puy de Dome research station after a night inside cloud.

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